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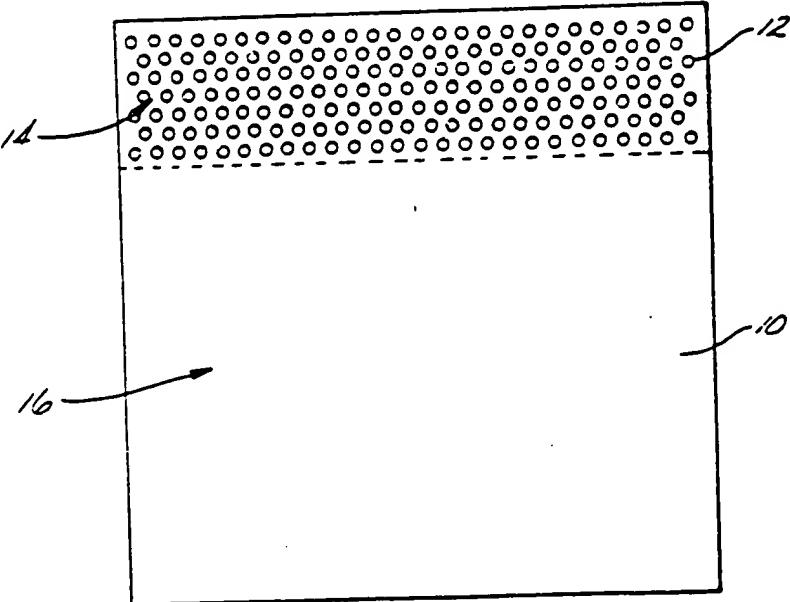
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## (54) Title: REMOVABLE LABELS

## (57) Abstract

Removable label stock and a process for its preparation. The stock (10) comprises discontinuous emulsion adhesive segments (12) on at least a portion (14) of one side of a face thereof. The amount of adhesive present for contact with a smooth surface is from about 10 percent to about 30 percent of the amount of adhesive which would have occupied the portion if the face stock were continuously coated with the adhesive. The emulsion adhesive segments have an average height, relative to the surface of the face stock, of from about 15 to about 35 microns; a room-temperature storage modulus of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians per second, a room-temperature storage modulus of from about  $4 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians per second, a room-temperature storage modulus of from about  $1 \times 10^4$  to about  $8 \times 10^4$  Pas-



cal at a deformation rate of about  $10^{-2}$  radian per second, and a room-temperature storage modulus of from about  $6 \times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate of about  $10^{-3}$  radian per second; a 180° peel strength on a stainless steel panel, having a roughness of about 0.5 micro-inch, of from about 0.5 pound to about 2 pounds per inch at a coating weight of about 40 grams/m<sup>2</sup>; and a glass-transition temperature of less than 250° Kelvin.

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15 REMOVABLE LABELS

Cross Reference to Related Application

This is a continuation-in-part of Application  
Serial No. 599,628, filed April 12, 1984.

20 Background of the Invention

The printing of discontinuous patterns of adhesive  
is old in the art. Patents which may be mentioned are  
U.S. Patents 1,882,593; 2,191,704; 2,515,423; 2,822,290;  
2,607,711; 3,311,489; 3,508,947; 3,268,357; 2,721,810;  
3,505,497; 3,627,559; 3,940,868; 3,174,888; 3,741,786;  
and 3,857,931. Reasons for printing such adhesive pat-  
terns have varied widely.

25 Post-it™, a product manufactured and sold by 3M  
Company, has enjoyed success as a removable notepaper.  
As currently understood, Post-it® is constructed by  
applying on a primed side of a paper stock, adhesive  
microspheres, possibly prepared according to U.S. Patent  
3,691,140 to Silver and/or U.S. Patent 4,166,152 to Baker,  
30 in a random fashion. This provides a surface which is  
coated with adhesive microspheres. The adhesive micro-  
spheres provide an array of peaks and valleys which in  
turn provide, in a microscopic sense, a generally random,  
discontinuous pattern. Examination has suggested the

-2-

1 product to have an effective height differential of about  
20 microns, with approximately 37 percent of the adhesive  
surface being available for contact with a smooth sub-  
strate. Contact is initiated at the peaks of adhesive  
5 microspheres.

It would be desirable to have greater flexibility in  
the construction of removable label stock to meet a wide  
variety of applications.

10 Summary of the Invention

According to the present invention, there is provided  
removable label stock comprising a plurality of discontin-  
uous, adhesive segments in a pattern on at least a portion  
of at least one side of the carrier face stock and present  
15 in an amount to provide, in the zone bounded or defined by  
the adhesive, from about 10 percent to about 30 percent,  
preferably from about 15 percent to about 25 percent,  
of the adhesive present in the pattern, available for  
contact with a smooth substrate, such as stainless steel  
20 or glass, relative to the amount of adhesive which would  
have been present in the pattern if the adhesive were  
applied as a continuous film. To achieve this level for  
effective contact, from about 30 percent to about 75 per-  
cent of the zone which would have been occupied by the  
25 continuous film, is covered by adhesive segments. The  
adhesive segments are formed from an aqueous emulsion  
of particles which are about 1 micron or less in diameter  
and which yield, by water evaporation and/or, in the case  
of paper stock, by stock penetration, adhesive segments  
30 formed by agglomeration of the particles. The segments  
have an average height of from about 15 to about 35  
microns, preferably from about 20 to about 35 microns.  
The width of segments is in the range of from about 50 to  
about 400 microns, preferably from about 100 to about 300  
35 microns.

-3-

Adhesive properties are critical to removability, and may be controlled by in-situ and/or post-polymerization crosslinking. Properties determined to be necessary are, that the adhesive have a room-temperature storage modulus ( $G'$ ) of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians/second; from about  $4 \times 10^4$  to about  $5 \times 10^5$ , preferably about from about  $5 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians/second; from about  $1 \times 10^4$  to about  $8 \times 10^4$ , preferably from about  $1 \times 10^4$  to about  $6 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$  radian/second; and from about  $6 \times 10^3$  to about  $7 \times 10^4$ , preferably from about  $6 \times 10^3$  to about  $5 \times 10^4$  Pascal at a deformation rate of about  $10^{-3}$  radian/second, as determined by ASTM D-4065-82, modified for soft matrices.

The adhesive is further characterized as providing a  $180^\circ$  peel strength on the stainless steel panel, having a roughness of 0.5 micro-inch, of from about 0.5 pound to about 2 pounds per inch, at a coating weight of 40 grams/meter<sup>2</sup>. The effective average height of the adhesive segments at from about 15 to about 35 microns, is essential for broad-based application of a removable product, taking into account roughness of the carrier stock and substrate surface. Width of the adhesive segments is important to prevent fiber-picking of a paper substrate.

The adhesive employed further has a glass-transition temperature of less than about  $250^\circ$  Kelvin, preferably from about  $200^\circ$  to about  $235^\circ$  Kelvin, and, generally, a molecular weight of greater than two times the entanglement molecular-weight, preferably greater than about ten times the entanglement molecular-weight.

- 4 -

1        Although a wide variety of adhesives may be employed  
2        to construct removable stock, it is presently preferred to  
3        employ an acrylic-polymer adhesive printed from an emul-  
4        sion by a gravure cylinder onto the face stock. Acrylic  
5        adhesive will not stain paper nor vinyl substrates. Where  
6        the face material is paper, it is preferred that the  
7        solids content of the adhesive be at least 60 percent by  
8        weight and have a gel content of from about 65 percent to  
9        about 90 percent, preferably from about 70 percent to  
10      about 90 percent by weight, on a dry basis.

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Brief Description of the Drawings

Attached FIGS. 1 and 2 illustrate, in somewhat exaggerated terms, the generalized structure of notepaper having a discontinuous, removable adhesive gravure-printed thereon.

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Detailed Description

5       The present invention is directed to providing removable, pressure-sensitive-adhesive stock having adhesive segments contained thereon, as provided by gravure-printing, screen-printing, or the like.

10      With reference now to FIGS. 1 and 2, the face stock 10 of a depicted notepaper has printed thereon an array of discontinuous adhesive segments 12 which occupy a portion, zone 14, of the total area (zone 14 + zone 16) 15      of the illustrated surface of the stock. In respect of zone 14, the amount of adhesive applied occupies from about 30 percent to about 75 percent of the area which would have been occupied by adhesive if the adhesive had been continuously coated over zone 14.

20      With particular reference to FIG. 2, only a portion of the applied adhesive may be available for contact with a substrate unless, through compression of the adhesive, the area of contact is increased. The area available for contact to a smooth surface, such as stainless steel or glass, by a portion of given segment 12, depicted as 18, is from about 10 percent to about 30 percent of the total area of zone 14, normally from about 15 percent to about 25 percent. The actual amount of contact area will 25      vary depending upon the roughness or smoothness of the face stock and the roughness or smoothness of the substrate. As illustrated in FIG. 2, which microscopically depicts a paper surface, the valleys of the paper may preclude contact by making a segment inaccessible for contact. By contrast, more or less contact may occur 30      with another rough surface, such contact due to nestling or a lack thereof. To account for wide variations in face stock and substrate, height of the adhesive segments employed should be, on the average, from about 15 to about 35 microns, preferably from about 20 to about 35 microns,

-7-

more preferably, about 30 microns. Adhesives employed are emulsion adhesives. Acrylic polymers are preferred. By the term "acrylic polymer" there is meant homopolymers and, preferably, copolymers containing at least one unsaturated monomer such as acrylic acid, butylacrylate, 2-ethylhexylacrylate, methacrylic acid, isoctylacrylate and the like, for the formation of acrylic polymer systems. Acrylic polymer systems are preferred, as they do not stain paper nor vinyl. Rubber-based emulsion polymers and the like, having viscoelastic properties as recited herein, may also be used where staining is not critical. The net adhesive available on a substrate is characterized as having a glass-transition temperature of less than about 250° Kelvin, preferably from about 200° to about 235° Kelvin, and a molecular weight greater than two times the entanglement molecular-weight, preferably at least ten times the entanglement molecular-weight. Gel, as induced by crosslinking, should be from about 65 percent to about 90 percent, preferably from about 70 percent to about 90 percent by weight, on a dry basis. Gel content is the amount of polymer insoluble in tetrahydrofuran.

While there is sufficient tenacity for the adhesive to remain on the face stock, to enable removability from the substrate the adhesive has a storage modulus ( $G'$ ) of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$ , preferably from about  $3 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians/second; from about  $4 \times 10^4$  to about  $5 \times 10^5$ , preferably from about  $5 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians/second; from about  $1 \times 10^4$  to about  $8 \times 10^4$ , preferably from about  $1 \times 10^4$  to about  $6 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$  radian/second; and from about  $6 \times 10^3$  to about  $7 \times 10^4$ , preferably from

-8-

1      about  $6 \times 10^3$  to about  $5 \times 10^4$  Pascal at a deformation  
 rate of about  $10^{-3}$  radian/second, as determined by  
 ASTM D-4065-82, modified for soft matrices.

5      The adhesive is further characterized as having a  
 180° peel, as determined by ASTM D-3330-81, from a stain-  
 less steel panel having a surface roughness of about 0.5  
 micro-inch, and from about 0.5 pound to about 2 pounds per  
 inch when such adhesive is applied at a continuous-coating  
 weight of 40 grams/meter<sup>2</sup>, with 2-mil mylar as the face  
 stock.

10     As indicated, the adhesive is printed from an emul-  
 sion normally containing particles of about one micron or  
 less in diameter which, upon evaporation of water from the  
 emulsion, coalesce to form adhesive segments having an  
 average height of from about 15 to about 35 microns,  
 preferably from about 20 to about 35 microns. Running  
 together of the applied adhesive is to be avoided at all  
 times, as performance in this instance is likely the most  
 inferior because of loss of effective height of the formed  
 adhesive to substantially less than 15 microns.

15     Table I, below, shows the general composition and  
 properties of the preferred adhesives for use in the  
 practice of the instant invention.

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Table I

	<u>Adhesive A</u>	<u>Adhesive B</u>
180° Peel (lb/in) (40 g/m <sup>2</sup> )	1.0	0.99
Glass Transition Temperature, Tg	213°K	203°K
Deformation Rate, Radians/Second	Storage Modulus at about 25°C (G') Pascal (N/m <sup>2</sup> )	
$10^{+4}$	$4.5 \times 10^5$	$1.0 \times 10^6$
$10^{+2}$	$9.0 \times 10^4$	$1.7 \times 10^5$
$10^{-2}$	$1.1 \times 10^4$	$2.4 \times 10^4$
$10^{-3}$	$9.0 \times 10^3$	$1.4 \times 10^4$

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-9-

Adhesive A contains about 60 percent by weight 2-ethylhexylacrylate, about 38 percent by weight butyl-acrylate, and about 2 percent by weight methacrylic acid, crosslinked with about 0.385 percent by weight CX-100, an aziridine crosslinker, and about 1.15 percent by weight dioctylphthalate. Gel content is from about 70 percent to 80 percent.

Adhesive B contains about 93.5 percent by weight 2-ethylhexylacrylate, about 5 percent by weight acrylic acid, and about 1.5 percent by weight hexanedioldiacrylate as the crosslinker.

In creating an adhesive of the required characteristics, crosslinking is employed only to the extent necessary to reduce the aggressiveness of the adhesive. Crosslinking may occur in situ during polymerization or prior to or after printing of the adhesive on the face stock.

Adhesive segments applied to the face stock are, indeed, microscopic, and the invention will now be described in detail by means of the producing removable adhesive stock in accordance with the instant invention.

It is presently preferred to use gravure-printing from a gravure cylinder having cells of an effective mean diameter of from about 100 to about 400 microns, preferably from about 150 to about 300 microns, more preferably from about 200 to about 300 microns; with a cell depth ranging from about 25 to about 70 microns, preferably from about 50 to about 70 microns; generally spaced from each other by a distance of up to about 200 microns; and containing from about 50 to about 120 cells/inch, the number of cells provided being inversely proportional to cell diameter. Again, the cells are to enable coating of from about 30 percent to about 75 percent of the area of the face stock which would otherwise be continuously coated with the adhesive. Effective coating weight ranges from about 3 to about 10 grams/m<sup>2</sup>, preferably from about 4 to about 7 grams/m<sup>2</sup>.

-10-

1        Because of the microscopic nature of the cells, con-  
tained emulsion is removed by capillary action. Removal  
is incomplete, leading to printing of segments of irregu-  
lar shape. At their maximum width, the segments will  
5        range from about 50 to about 400 microns, preferably from  
about 100 to about 300 microns. The lower limit specified  
is that necessary for contact. The upper limit specified  
is that necessary to avoid paper-pick, i.e., picking of  
paper fibers from a paper substrate.

10      Face stock to which adhesive can be applied may  
range widely, depending upon the end-use application: from  
a smooth surface, such as mylar; a semi-smooth surface,  
such as metal film or primed or coated paper; or a rough  
15     surface, such as unprimed paper. When the face stock is  
paper, the area that is to be printed with the adhesive  
may be primed with a primer, such as a composition formed  
of about 80 percent by weight zinc oxide and about 20 per-  
cent by weight polymethylmethacrylate, to retard moisture  
penetration. Corona-treating the paper and/or pre-wetting  
20     the paper can also be used to enhance adhesive printing.  
These procedures enable tenacious adhesion to the paper,  
as water leaves the emulsion to form the adhesive seg-  
ments. Where corona-treating is used, an effective  
25     corona can be generated at voltages of about 350 volts or  
more and at currents of 7 amps or more, for paper moving  
at web speed of from 100 to 175 feet per minute. In  
addition, transfer of emulsion to substrate may be eased  
by creating an electrostatic differential between the  
cylinder and the substrate.

30      It is presently preferred to print on uncoated paper  
with an acrylic emulsion having a solids content of at  
least about 60 percent, the acrylic polymer having a gel  
content of from about 70 percent to about 90 percent by  
weight, on a dry basis.

-11-

1 Products produced in accordance with the instant  
invention, and depending upon the nature of the emulsion  
adhesive employed and the stock to which it is applied,  
can be used for a variety of applications. In addition  
5 to notepaper and removable tape for conventional office  
use, they may be employed as temporary bumper stickers,  
as temporary identification tags, and for a wide variety  
of commercial applications.

10 While nowise limiting, the following is an example of  
removable stock prepared in accordance with the instant  
invention.

Example

15 There was processed, canary yellow paper of a weight  
of 50 pounds per ream, a caliper of 4 mils, a Sheffield  
Smoothness Test of 250 T/W, Gurley Porosity of 15, Cobb  
Size of 22.3, and an ash content of from 10 to 12 percent.  
The paper was passed at a web speed of 175 feet per minute  
through a corona formed at 350 volts and 10 amps. An  
20 emulsion of Adhesive A was printed from a gravure cylinder,  
11.5 inches in diameter, with 50 percent of the surface  
covered with cells of 250 microns in diameter and having a  
depth of 50 microns. There was formed on the paper a band  
approximately 17mm wide. A pad was formed from layers of  
25 adhesive-coated paper. Following are the forces required  
to remove the adhesive from certain substrates by peeling  
across the substrate:

Table II

	<u>Substrate</u>	<u>Adhesion, gram-force/sheet</u>
30	Xerox® Bond	46
	Unprinted Magazine Paper	72
	Typing Bond	21
35	Rough Vinyl	15
	Smooth Vinyl	41

-12-

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Control

5 A sample removable note paper was prepared by applying through a 120-mesh screen, an emulsion of a polymer containing about 62 parts by weight 2-ethylhexyl acrylate, 18 parts by weight methyl methacrylate, 5 parts by weight acrylic acid, and 15 parts by weight dibutyl fumarate, onto 60-pound Kromekote paper, made by Champion International. There were formed adhesive segments having an 10 average width of about 100 microns and an average height of about 15 microns. The adhesive of the segments had a gel content of about 58 percent. The rheological properties are shown in Table III.

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Table III

	<u>Deformation Rate, radians/second</u>	<u>Storage Modulus at about 25°C Pascal (N/m<sup>2</sup>)</u>
	10 <sup>4</sup>	2.2 x 10 <sup>6</sup>
	10 <sup>2</sup>	2.1 x 10 <sup>5</sup>
20	10 <sup>-2</sup>	7.5 x 10 <sup>3</sup>
	10 <sup>-3</sup>	4 x 10 <sup>3</sup>

25 When the product was applied to stainless steel, with a dwell time of 20 minutes, there resulted 90° peel from the stainless steel at a peel speed of 300 inches per minute, which gave paper tear. The product was unsuitable as a removable label.

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The emulsion adhesives of the Example and the Control were applied through 40-, 60-, and 80-,mesh screens to the paper of the Example, to provide adhesive segments of respective average widths of 390, 310, and 250 microns. Samples of the construction were applied to

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-13-

a legal pad known as Padmaster, manufactured and sold by Bradner Central Co., of Chicago, Illinois. Performance at different rates of peel is shown in Table IV.

Table IV

<u>Adhesive Width</u>	<u>Rate of Peel in./min.</u>	<u>Example</u>	<u>Adhesive</u>	<u>Control</u>
.0	390	300	paper pick	paper tear
	390	88	clean removal	paper tear
15	310	300	slight paper pick	paper tear
	310	88	clean removal	paper tear
25	250	300	clean removal	paper pick
	250	88	clean removal	slight paper pick

The above Table establishes the importance of segment width and adhesive selection to proper label performance.  
A rate of peel of 300 inches per minute is the highest rate that could be anticipated for normal use of the product. The most frequent rate of peel would be in the order of from about 75 to about 150 inches per minute.

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-14-

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WHAT IS CLAIMED IS:

1. A removable label stock comprising a plurality  
5 of discontinuous emulsion adhesive segments on at least  
a portion of at least one side of a face stock and  
present in an amount to provide from about 10 to about 30  
percent of the adhesive available for contact with a  
smooth substrate in a zone formed by the emulsion adhesive  
10 segments relative to the amount of adhesive which would  
have occupied said zone if continuously coated with said  
adhesive, said emulsion adhesive segments having an  
average height relative to the surface of the face stock  
of from about 15 to about 35 microns; a room-temperature  
15 storage modulus of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$   
Pascal at a deformation rate of about  $10^4$  radians per  
second, a room-temperature storage modulus of from about  
4  $\times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of  
about  $10^2$  radians per second, a room-temperature storage  
20 modulus of from about  $1 \times 10^4$  to about  $8 \times 10^4$  Pascal  
at a deformation rate of about  $10^{-2}$  radian per second,  
and a room-temperature storage modulus of from about  
6  $\times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate of  
about  $10^{-3}$  radian per second; a 180° peel strength on a  
25 stainless steel panel, having a roughness of about 0.5  
micro-inch, of from about 0.5 pound to about 2 pounds  
per inch at a coating weight of about 40 grams/m<sup>2</sup>; and  
a glass-transition temperature of less than 250° Kelvin.

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-15-

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2. A removable label stock comprising a plurality  
of discontinuous emulsion adhesive segments on at least  
a portion of at least one side of a face stock and  
5 present in an amount to provide from about 1.0 to about 30  
percent of the adhesive available for contact with a  
smooth substrate in a zone formed by the emulsion adhesive  
segments relative to the amount of adhesive which would  
have occupied said zone if continuously coated with said  
10 adhesive, said emulsion adhesive segments having an  
average height relative to the surface of the face stock  
of from about 20 to about 35 microns; a room-temperature  
storage modulus of from about  $3 \times 10^5$  to about  $1.5 \times 10^6$   
15 Pascal at a deformation rate of about  $10^4$  radians per  
second, a room-temperature storage modulus of from about  
 $5 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of  
about  $10^2$  radians per second, a room-temperature storage  
modulus of from about  $1 \times 10^4$  to about  $6 \times 10^4$  Pascal  
at a deformation rate of about  $10^{-2}$  radian per second,  
20 and a room-temperature storage modulus of from about  
 $6 \times 10^3$  to about  $5 \times 10^4$  Pascal at a deformation rate of  
about  $10^{-3}$  radian per second; a 180° peel strength on a  
stainless steel panel, having a roughness of about 0.5  
25 micro-inch, of from about 0.5 pound to about 2 pounds  
per inch at a coating weight of about  $40 \text{ grams/m}^2$ ; and  
a glass-transition temperature of less than 250° Kelvin.

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-16-

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3. A removable label stock comprising a plurality  
of discontinuous emulsion adhesive segments having a  
maximum width of from about 50 to about 400 microns on  
5 at least a portion of at least one side of a paper  
face-stock and present in an amount to provide from about  
10 to about 30 percent of the adhesive available for  
contact with a smooth substrate in a zone formed by the  
emulsion adhesive segments relative to the amount of  
10 adhesive which would have occupied said zone if  
continuously coated with said adhesive, said emulsion  
adhesive segments having an average height relative to  
the surface of the paper face-stock of from about 15 to  
about 35 microns; a room-temperature storage modulus of  
15 from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a  
deformation rate of about  $10^4$  radians per second, a  
room-temperature storage modulus of from about  $4 \times 10^4$   
to about  $5 \times 10^5$  Pascal at a deformation rate of about  
10<sup>2</sup> radians per second, a room-temperature storage  
modulus of from about  $1 \times 10^4$  to about  $8 \times 10^4$  Pascal  
at a deformation rate of about  $10^{-2}$  radian per second,  
20 and a room-temperature storage modulus of from about  
6  $\times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate of  
about  $10^{-3}$  radian per second; and a 180° peel strength  
25 on a stainless steel panel, having a roughness of about  
0.5 micro-inch, of from about 0.5 pound to about 2 pounds  
per inch at a coating weight of about 40 grams/m<sup>2</sup>, said  
adhesive segments being formed of an acrylic adhesive  
polymer having a glass-transition temperature of less  
30 than 250° Kelvin.

-17-

4. A removable label stock comprising a plurality of discontinuous emulsion adhesive segments having a maximum width of from about 50 to about 400 microns on at least a portion of at least one side of a paper face-stock and present in an amount to provide from about 10 to about 30 percent of the adhesive available for contact with a smooth substrate in a zone formed by the emulsion adhesive segments relative to the amount of adhesive which would have occupied said zone if continuously coated with said adhesive, said emulsion adhesive segments having an average height relative to the surface of the paper face-stock of from about 20 to about 35 microns; a room-temperature storage modulus of from about  $3 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians per second, a room-temperature storage modulus of from about  $5 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians per second, a room-temperature storage modulus of from about  $1 \times 10^4$  to about  $6 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$  radian per second, and a room-temperature storage modulus of from about  $6 \times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate of about  $10^{-3}$  radian per second; and a  $180^\circ$  peel strength on a stainless steel panel, having a roughness of about 0.5 micro-inch, of from about 0.5 pound to about 2 pounds per inch at a coating weight of about 40 grams/m<sup>2</sup>, said adhesive segments being formed of an acrylic adhesive polymer having a gel content of from about 65 percent to about 90 percent, and a glass-transition temperature of less than than 250° Kelvin.

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5. A removable label stock as claimed in any one of claims 1 to 4 in which the adhesive has a glass-transition temperature of from about 200° to about 235°

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Kelvin.

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6. A removable label stock as claimed in any one of claims 1 to 5 in which the adhesive is formed of an acrylic polymer containing an interpolymerized amount of at least one monomer selected from the group consisting of acrylic acid, butylacrylate, 2-ethylhexyl acrylate, methacrylic acid, and isoctyl acrylate.

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7. A removable label stock as claimed in any one of claims 1 to 5 in which the adhesive is an acrylic polymer comprising, based on the weight of the monomers, about 60 percent by weight 2-ethylhexyl acrylate, about 38 percent by weight butyl acrylate, and about 2 percent by weight methacrylic acid, crosslinked to a gel content of from about 65 percent to about 90 percent by weight, on a dry basis.

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8. A removable label stock as claimed in any one of claims 1 to 7 in which from about 15 to about 25 percent of the adhesive in the zone is available for contact with a smooth surface, relative to the amount of adhesive in the zone which would have been available for contact with the smooth surface if said zone were continuously coated with said adhesive.

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9. A removable label stock as claimed in any one of claims 1 to 8 in which the adhesive segments have an average width of from about 200 to about 300 microns.

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-19-

10. A removable label stock as claimed in any one of claims 1 to 9 in which the adhesive segments provide contact to a smooth substrate of from about 10 percent to about 30 percent of the zone of the label stock in which the adhesive segments are contained.

11. A process for the production of a removable label stock, which process comprises applying from a gravure cylinder having an array of cells of a mean diameter of from about 100 to about 400 microns and a cell depth of from about 50 to about 70 microns, onto at least a portion of one surface of a face stock, aqueous-emulsion adhesive segments in a discontinuous pattern, and forming from the applied emulsion adhesive segments having an average height, relative to the face stock, of from about 15 to about 35 microns and being present to provide for contact with a smooth substrate, from about 10 percent to about 30 percent of the adhesive which would have been available for contact if said portion of the face stock were continuously coated with said adhesive, said adhesive having a room-temperatuire storage modulus of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians per second, a room-temperature storage modulus of from about  $4 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians per second, a room-temperature storage modulus of from about  $1 \times 10^4$  to about  $8 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$  radian per second, and a room-temperature storage modulus of from about  $6 \times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate of about  $10^{-3}$  radian per second; a 180° peel strength on a stainless steel panel, having a roughness of about 0.5 micro-inch, of from about 0.5 pound to about 2 pounds per inch at a coating weight of 40 grams/m<sup>2</sup>; and a glass-transition temperature of less than 250° Kelvin.

-20-

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12. A process as claimed in claim 11 in which the adhesive emulsion is applied from a gravure cylinder having an array of cells of a mean diameter of from about 200 to about 400 microns, and in which there are formed from the applied emulsion, adhesive segments having a height, relative to the face stock, of from about 20 to about 35 microns and being present to provide for contact with a smooth substrate, said adhesive segments having a room-temperature storage modulus of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of about  $10^4$  radians per second, a room-temperature storage modulus of from about  $4 \times 10^4$  to about  $5 \times 10^5$  Pascal at a deformation rate of about  $10^2$  radians per second, a room-temperature storage modulus of from about  $1 \times 10^4$  to about  $6 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$  radian per second, and a room-temperature storage modulus of from about  $6 \times 10^3$  to about  $5 \times 10^4$  Pascal at a deformation rate of about  $10^{-3}$  per second; and a 180° peel strength on a stainless steel panel, having a roughness of about 0.5 micro-inch, of from about 0.15 pound to about 2 pounds per inch at a coating weight of 40 grams/m<sup>2</sup>.

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13. A process for the production of a removable label stock, which process comprises applying from a gravure cylinder having an array of cells of a mean diameter of from about 100 to about 400 microns and a cell depth of from about 25 to about 70 microns, onto at least a portion of one surface of a paper face-stock, aqueous acrylic-emulsion adhesive segments in a discontinuous pattern and forming from the applied acrylic emulsion, acrylic-adhesive segments having an average height, relative to the face stock, of from about 15 to about 35 microns and being present to provide for contact with a smooth substrate, from about 10 percent to

-21-

Claim 13, continued. . .  
about 30 percent of the adhesive which would have been  
available for contact if said portion of the face stock  
were continuously coated with said adhesive, said  
adhesive segments having a room-temperature storage  
modulus of from about  $2 \times 10^5$  to about  $1.5 \times 10^6$  Pascal  
at a deformation rate of about  $10^4$  radians per second,  
a room-temperature storage modulus of from about  $4 \times 10^4$   
to about  $5 \times 10^5$  Pascal at a deformation rate of about  
 $10^2$  radians per second, a room-temperature storage  
modulus of from about  $1 \times 10^4$  to about  $8 \times 10^4$  Pascal  
at a deformation rate of about  $10^{-2}$  radian per second,  
and a room-temperature storage modulus of from about  
 $6 \times 10^3$  to about  $7 \times 10^4$  Pascal at a deformation rate  
of about  $10^{-3}$  radian per second; and a  $180^\circ$  peel strength  
on a stainless steel panel, having a roughness of about  
0.5 micro-inch, of from about 0.15 pound to about 2  
pounds per inch at a coating weight of 40 grams/m<sup>2</sup>, said  
acrylic adhesive having a glass-transition temperature  
of less than 250° Kelvin.

14. A process as claimed in claim 13 in which the  
acrylic emulsion is applied from a gravure cylinder  
having an array of cells of a mean diameter of from about  
200 to about 400 microns, and in which there is formed  
from the applied acrylic emulsion, adhesive segments  
having an average height, relative to the face stock, of  
from about 20 to about 35 microns, said adhesive segments  
having a room-temperature storage modulus of from about  
 $3 \times 10^5$  to about  $1.5 \times 10^6$  Pascal at a deformation rate of  
about  $10^4$  radians per second, a room-temperature storage  
modulus of from about  $5 \times 10^4$  to about  $5 \times 10^5$  Pascal at  
a deformation rate of about  $10^2$  radians per second, a  
room-temperature storage modulus of from about  $1 \times 10^4$  to  
about  $6 \times 10^4$  Pascal at a deformation rate of about  $10^{-2}$   
radian per second, and a room-temperature storage modulus

-22-

1       Claim 14, continued. . .  
of from about  $6 \times 10^3$  to about  $5 \times 10^4$  Pascal at a  
deformation rate of about  $10^{-3}$  radian per second; and a  
5       180° peel strength on a stainless steel panel, having a  
roughness of about 0.5 micro-inch, of from about 0.15  
pound to about 2 pounds per inch at a coating weight of  
40 grams/m<sup>2</sup>.

10       15. A process as claimed in any one of claims 11 to  
14 in which the adhesive emulsion has a glass-transition  
temperature of from about 200° to about 235° Kelvin.

15       16. A process as claimed in any one of claims 11 to  
15 in which from about 15 percent to about 25 percent of  
the adhesive is available for contact with a smooth  
surface, relative to the amount of adhesive in the  
portion for contact with the smooth surface if said  
portion were continuously coated with adhesive.

20       17. A process as claimed in any one of claims 11 to  
16 in which the paper face-stock is coated with a primer.

25       18. A process as claimed in any one of claims 11 to  
17 in which the emulsion has a solids content of at least  
60 percent by weight and the adhesive has a gel content  
of from about 65 percent to about 90 percent, on a dry  
basis.

30       19. A process as claimed in any one of claims 11 to  
18 in which the adhesive is formed of an acrylic polymer  
containing an interpolymerized amount of at least one  
monomer selected from the group consisting of acrylic  
acid, butylacrylate, 2-ethylhexyl acrylate, methacrylic  
acid, and isooctyl acrylate.

-23-

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20. A process as claimed in any one of claims 11 to 18 in which the adhesive is an acrylic polymer comprising, based on the weight of the monomers, about 60 percent by weight 2-ethylhexyl acrylate, about 38 percent by weight butyl acrylate, and about 2 percent by weight methacrylic acid, crosslinked to a gel content of from about 65 percent to about 90 percent by weight, on a dry basis.

21. A process as claimed in any one of claims 11 to 20 in which the adhesive segments have an average maximum width of from about 50 to about 400 microns.

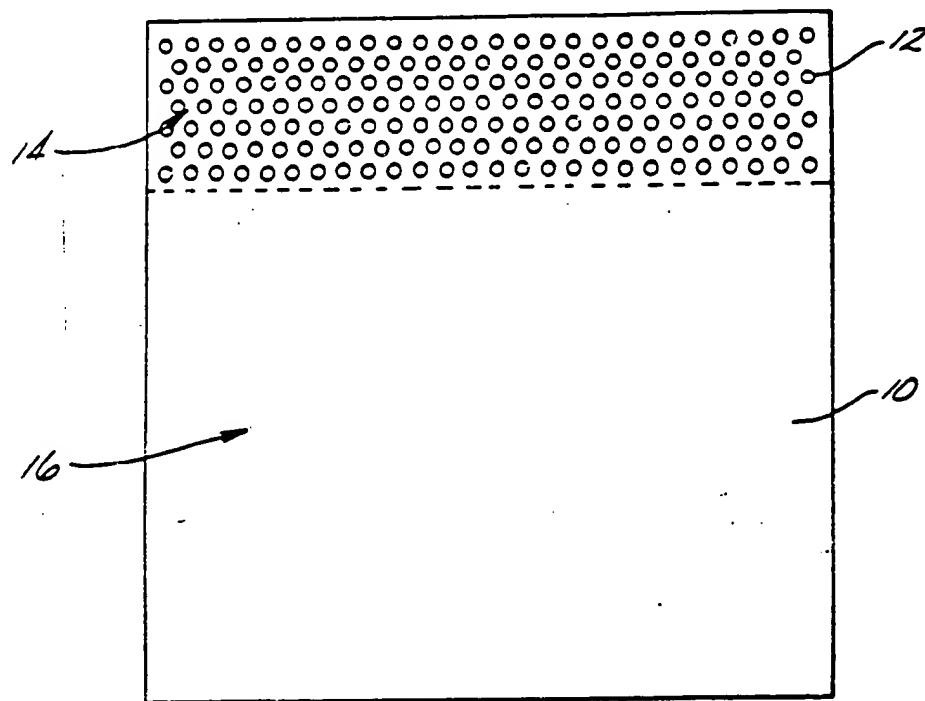
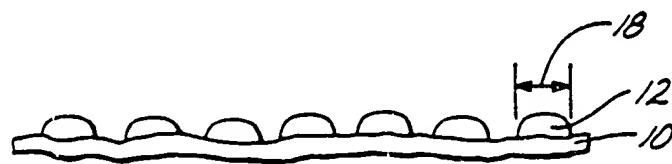
22. A process as claimed in any one of claims 11 to 21 in which the adhesive segments have an average maximum width of from about 50 to about 300 microns.

23. A process as claimed in any one of claims 11 to 22 in which the adhesive segments provide contact to a smooth substrate of from about 10 percent to about 30 percent of the zone of the label stock in which the adhesive segments are contained.

25. A process as claimed in any one of claims 11 to 23 in which the adhesive segments are applied to a coating weight of from about 3 to about 10 grams per square meter, on a dry basis.

26. A process as claimed in any one of claims 11 to 23 in which the adhesive segments are applied to a coating weight of from about 4 to about 7 grams per square meter, on a dry basis.

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*Fig.1.**Fig.2.*

## INTERNATIONAL SEARCH REPORT

International Application No PCT/US85/00602

## I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) \*

According to International Patent Classification (IPC) or to both National Classification and IPC  
 IPC<sup>3</sup> B05D 5/10, B32B 3/10, B32B 7/06, B32B 7/14, B32B 7/10  
 U.S. 428/194, 198, 202; 427/208.6, 256, 261

## II. FIELDS SEARCHED

Minimum Documentation Searched \*

Classification System	Classification Symbols
U.S.	428/40, 194, 198, 202 427/208.6, 256, 261

Documentation Searched other than Minimum Documentation  
to the Extent that such Documents are Included in the Fields Searched \*

## III. DOCUMENTS CONSIDERED TO BE RELEVANT \*\*

Category *	Citation of Document, * with indication, where appropriate, of the relevant passages **	Relevant to Claim No. **
X	US, A, 3,033,702 PUBLISHED 05 MAY 1962 FENSELAU	1-14
X	US, A, 4,460,634 PUBLISHED 07 JULY 1984 HASEGAWA	1-14
X	US, A, 2,264,628 PUBLISHED 12 DECEMBER 1941 ENGERT ET AL	1-14

\* Special categories of cited documents: \*\*

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s); or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"A" document member of the same patent family

## IV. CERTIFICATION

Date of the Actual Completion of the International Search \*

26 APRIL 1985

Date of Mailing of this International Search Report \*

09 MAY 1985

International Searching Authority \*

ISA/US

Signature of Authorized Officer \*\*  
*Alexander S. Thomas*  
 ALEXANDER S. THOMAS